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Density Changes in Plutonium Observed from Accelerated Aging using Pu-238 Enrichment

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Abstract

In support of Stockpile Stewardship activities, accelerated aging tests on a plutonium alloy enriched with 7.3 atomic percentage of ^{238}Pu is underway using dilatometry at 35, 50, and 65°C and immersion density measurements of material stored at 50°C. Changes in density are expected from radiation damage in the lattice and helium in-growth. After twenty-five equivalent years of aging, the dilatometry data shows that the alloys at 35°C have expanded in volume by 0.11% to 0.12% and have started to exhibit a near linear expansion behavior primarily caused by the helium accumulation. The average He-to-vacancy ratio from tested specimens was determined to be around 2.3. The model for the lattice damage and helium in-growth accurately represents the volume swelling at 35°C. The density converted from the dilatometry corresponds well to the decreasing density trend of reference plutonium alloys as a function of time.

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1. Introduction

The assessment of aging effects in the plutonium lies at the heart of science-based stockpile stewardship [1, 2]. Plutonium, because of its radioactive nature, ages by means of self-irradiation damage and thus produces Frenkel-type defects (vacancies and self-interstitial atoms) and defect clusters. The half-life of the major isotope ^{239}Pu is 24,390 years, and it decays mainly by α -particle decay. Each decay event produces two energetic nuclear particles, with the helium nucleus receiving just above 5 MeV and a recoil uranium nucleus receiving about 85 keV [1,2]. The α -particle captures two electrons from the plutonium metal to become helium in the lattice, which can associate with crystal defects such as vacancies. In principle, both the uranium and helium decay products can damage the crystal lattice through collision with plutonium atoms. However, the highly energetic α -particle loses nearly 99.9% percent of its energy to electrons, only heating the plutonium crystal lattice. In contrast, the heavier uranium nucleus converts nearly 75% of its 85 keV of decay energy into the atomic displacement, causing most of the initial damage to the plutonium lattice. Together, both α -particle and uranium nuclei produce roughly 2500 Frenkel-pairs for each radioactive decay event [1, 2]. While most of the initial damage is annealed out at room temperature, the remaining defects resulting from the residual lattice damage and helium formation drive microstructural and physical property changes. As a result, radiation damage from alpha decay in plutonium occurs at a rate of ~ 0.1 dpa (displacement per atom) per year, while the rate of helium production is ~ 41.1 appm (atomic parts per million) per year [1].

It is well known in the nuclear-energy industry that reactor steels undergo microstructural changes from radiation exposure, which include reduction in density, hardening, and embrittlement in reactor steels [3]. A typical void swelling behavior is characterized by three stages [1, 3]: an incubation period, a transient period, and a steady-state swelling period. The first stage is an incubation period during which void nucleation takes place with little or no change in initial density or volume. The second stage is a transient period during which an optimum ratio is established for the densities of voids and dislocations. The third stage is a steady-state swelling period where the volume swelling rate can accelerate to a value of 1% per dpa for irradiated alloys in reactor environments [3]. The duration of both incubation and transient periods commonly depends on temperature, dose rate, and initial material characteristics like composition and dislocation density [1, 3]. While these general features of swelling are observed in metals and alloys under prolonged exposure at high radiation dose and in the appropriate temperature range, these phenomena have not yet been observed in naturally aged plutonium alloys [1].

Limited prior research has been performed regarding plutonium aging at ambient temperature. The X-ray diffraction study by Chebotarev and Utkina on Pu alloys aged at room temperature for 2.5 years showed lattice expansion and the lattice parameter reached essentially a constant value in 2 to 3 years [4]. Positron annihilation measurements indicate that helium-filled vacancies are present in significant numbers in aged plutonium samples [5, 6]. Recent transmission electron microscopy investigation of aged Pu-Ga alloys revealed the presence of nanometer-sized helium bubbles formed from helium in-growth [7]. However, most experiments on self-irradiation damage in

plutonium (for example, resistivity measurements [2, 8, 9]) have been conducted at cryogenic temperatures over very short time periods where very little annealing occurs and do not represent the plutonium aging behavior at ambient temperature.

A preliminary assessment of radiation damage in gallium stabilized δ -plutonium at ambient temperature by W. Wolfer indicates the steady-state swelling or void swelling is expected to begin after 10 to 100 years after the fabrication [1]. This estimate is made by assuming that δ -plutonium has radiation and helium effects consistent with typical fcc metals. However, there is no evidence for void swelling in naturally aged plutonium alloys even after 42 years [7]. Clearly, experiments are needed to replace the current estimate with actual data on radiation-induced dimensional changes in plutonium.

Because effects over decades are of interest, a proposed approach is to accelerate the aging (decay) of plutonium as well as perform detailed characterization of new and aged materials. Because the α -decay rate of ^{238}Pu is nearly 300 times faster than that of ^{239}Pu , the rate of radiation damage can be accelerated. Thus, it is possible to evaluate extended-age plutonium (up to 60-years equivalent and possibly beyond) within a few years by doping plutonium with small fractions of ^{238}Pu . The resulting damage processes and primary defects with the doping ^{238}Pu will be unchanged compared to the plutonium alloy based on ^{239}Pu . With this approach, the acceleration of radiation damage must be matched by acceleration of the ensuing annealing of that damage as in naturally aged plutonium at ambient temperature. This ensuing acceleration can be achieved by raising the storage (aging) temperature. In this paper, we report on the initial volume changes observed from the in-situ dilatometry on the 7.3 at. % ^{238}Pu enriched plutonium alloy and supplemented by immersion density measurements on the 7.3 at. % ^{238}Pu enriched

plutonium and reference plutonium alloys of various ages stored at 50°C. In this work, the immersion density results are used to validate the dilatometry. Because of uncertainties in the proper annealing temperature to balance the accelerated damage, the dilatometry experiments are performed at three different temperatures, i.e., at 35°C, 50°C, and 65°C.

2. Experimental

2.1 Sample preparation and Chemical Analysis

Pyrochemical processing methods have been used to produce both reference and 7.3 at. % ^{238}Pu -enriched plutonium alloys with nominal compositions of ~2 at. % Ga. Details of processes are presented elsewhere [10]. Processes included direct oxide reduction of PuO_2 with Ca, electrorefining, alloying, and casting. Density and dilatometer specimens were machined from the alloy after annealing at ~450°C for 12 hours. Density specimens were cut into rectangular shape, weighing nominally 5 grams. Additional density specimens were made from two separate sources of chemically identical Pu-Ga alloys of ages 2.5 years (33 grams) and 22 years (1 and 3 grams). Dilatometer specimens were made into 2 mm square by 2 or 3 cm long with masses of 1.2 gram and 1.7 gram, respectively.

Isotopic and elemental analyses were carried out by Inductively Coupled Mass Spectrometry (ICP-MS) on the enriched alloy produced here and compared with the reference alloy (see Table 1). The analyses on the individual disks showed very small variations from the values in Table 1, as indicated by the standard deviations. Note that analysis for C, N, and Si could not be conducted by ICP-MS.

2.2 Dilatometry

Specially designed dilatometers have been set up inside a nitrogen atmosphere glovebox to monitor long-term growth resulting from the lattice damage and helium ingrowth in (7.3 at. %) ^{238}Pu -enriched alloys. The schematic of the dilatometer system is presented elsewhere [11]. Each dilatometer unit consists of a sample vacuum chamber fitted with three linear variable differential transducers (LVDTs). An LVDT measures minute changes, less than $0.02\text{ }\mu\text{m}$, in the position of a push-rod by monitoring changes in the inductance of a detector coil. In this system, the detector coil is placed outside of the sample chamber. Each dilatometer has three wells in a copper sample holder: one for a 3 cm long specimen, one for a 2 cm long specimen, and one for a 3 cm long system reference material (Schott glass “Zerodur”, “zero” expansion glass). Each dilatometer unit can hold two specimens of either circular or square cross-section, 2 or 3 cm long by 2 to 2.5 mm in diameter. Two different lengths of ^{238}Pu -enriched alloy specimens are used to differentiate between surface effects and volumetric swelling in the materials. Bulk changes are expected to be proportional to the length and any surface effects are expected to be nominally the same between the two samples. Plutonium specimens are placed in the temperature controlled copper sample holder that rests on an ultra-low expansion glass (Schott glass “Zerodur”) inside the sample chamber. This “Zerodur” provides a stable reference for length measurements, and its position is monitored continuously with one of the three LVDT rods. The other two LVDT rods are placed on the top ends of two plutonium specimens, which protrude from each well. The bottom ends of plutonium specimens rest on the “Zerodur” reference. Once the specimens and

the LVDT rods are positioned, the chamber is sealed with copper gaskets, evacuated with an oil-free vacuum pump and backfilled with high purity helium. The high purity helium in the chamber prevents any temperature gradient as a result of the specimen extending above the copper sample holder. The glovebox atmosphere is nitrogen, and it is maintained to within $\pm 0.5^\circ\text{C}$. The temperature of the dilatometer and its amplifier are maintained to within $\pm 0.1^\circ\text{C}$ by cooling water. With the temperature control and use of the “Zerodur” reference, the stability of the measurement over a 120 day period is better than $\pm 0.10\text{ }\mu\text{m}$, which is the systematic uncertainty in the measurement.

For the test specimens, three 2 cm and three 3 cm long dilatometer specimens of ^{238}Pu -enriched alloys were heat treated in high purity helium atmosphere for 30 minutes at 200°C , and shortly thereafter a pair of 2 and 3 cm length specimens was loaded into each dilatometer. The heat treatment was needed prior to the loading to anneal out the existing lattice damage formed during storage after specimen fabrication. Each system was then maintained at a separate temperature, i.e., at 35°C , 50°C , or 65°C .

2.3 Immersion Density

The immersion density equipment closely matches a design used by Bowman *et al.* [12] and uses about 200 ml of Fluorinert Electronic Liquid FC-43 as the immersion fluid. Prior to a measurement, the system is calibrated using NIST glass (SRM 1827A) and a tantalum specimen as standards. During the measurement process, dry weights of each specimen are measured to $\pm 0.01\text{ mg}$, giving a sensitivity of about $\pm 10^{-4}\text{ g/cm}^3$ and overall accuracy of about $\pm 10^{-3}\text{ g/cm}^3$. Densities of reference plutonium alloys of various ages are measured at ambient temperature. A correction needs to be applied to

the measured density to compensate for the heat generated by the alpha decay of plutonium. To get the actual density, the measured data needs to be corrected for the convection currents caused by the self heating of plutonium alloys in the FC-43 immersion fluid. In addition, because the ^{238}Pu -enriched alloys generate significantly more heat compared to the reference specimens, a test sample needs to be left overnight in the immersion bath to allow the temperature of the bath to stabilize and the measurements to be reproducible. As an example, a 5 g sample of 7.3 at. % ^{238}Pu -enriched alloy produces 225 mW of power. To correct for the convection current and obtain the actual density, a series of power generating samples were made of tantalum heated with tungsten heating wire, and heat inputs ranging from 0 to 1030 mW were run in the immersion density unit. The offset in density was fit to the following equation:

$$D_{\text{offset}} = 0.06839 \ln[1 + 0.006173Q] \quad (1)$$

where D_{offset} is the offset in density in units of g/cm^3 and Q is the heat input in mW. At 225 mW, a correction of $+0.06 \text{ g/cm}^3$ is required to convert the measured density into the actual density.

3. Results and Discussion

3.1 Dilatometry

The volume change (ΔV) normalized with the initial volume (V_0) of each enriched alloy sample at 35°C , 50°C , and 65°C is shown in Figure 1. The time is represented as an equivalent time (in years) calculated by taking the actual measurement time and multiplying by the accelerating factor (e.g. an initial factor of 18.16) which is determined from the difference in the activity between the reference alloy and ^{238}Pu -enriched alloy.

This accelerating factor will decrease as the material ages due primarily to decreasing concentration of ^{238}Pu in the specimen. Each dilatometer contains a long (3 cm) and short (2 cm) specimen. Since the volume expansion due to self-irradiation damage is assumed isotropic in the bulk of material and is small compared to the total volume, the $\Delta V/V_0$ of the specimen can be obtained with the approximate relation $\Delta V/V_0 \cong 3 \Delta L/L_0$ where $\Delta L/L_0$ represents the measured specimen length change (ΔL) normalized with the initial length (L_0). Table 2 shows the specimen length change (ΔL) and the volume expansion for each specimen after twenty-five equivalent years of aging. As plotted in Figure 1, all the enriched alloys have increased in volume significantly. There is a trend in $\Delta V/V_0$ curves with the temperature: samples expanded more with an increase in the test temperature. The specimens at 35°C have expanded in volume by 0.11 % to 0.12 % while the 2 cm specimen at 65°C has expanded by 0.22 %. Additionally, the 2 cm length specimens at 65°C and 50°C give higher volume change values compared to the 3 cm length specimens. The difference in $\Delta V/V_0$ values for 2 cm and 3 cm length specimens at 50°C and 65°C is believed to result mainly from surface effects (i.e. oxidation and/or interfacial reaction with dilatometry components) in the specimens. At 35°C, both 2 and 3 cm length specimens (open and closed squares in Figure 1) show similar $\Delta V/V_0$ values indicating minimal surface contribution, and the measured volume change is therefore representative of the bulk material. For comparison, Chebotarev and Utkina, using an X-ray diffraction technique, measured lattice expansion of Pu-Ga alloys aged at room temperature for 2.5 years [4]. They extrapolated their results to ~2 at. % Ga and obtained an approximate expansion value of $\Delta a/a_0 \approx 4.5 \times 10^{-4}$ or $\Delta V/V_0 \approx 1.35 \times 10^{-3}$ where a is a lattice constant. This expansion value is between the volume expansion of 65°C (2 cm)

and 65°C (3 cm) specimens at 2.5 years. Their results show that the lattice parameter of the Pu-Ga alloy increases as a result of prolonged self-irradiation at room temperature. The accumulation of vacancy and self-interstitial lattice defects from the initial cascade damage lead to increase in lattice parameters, which saturates in approximately two to three years.

3.2 Surface effects and volumetric swelling

The difference in the volume change between 2 and 3 cm length specimens at 50°C and 65°C is potentially due to surface effects. If only the bulk swelling accrues from the radiation damage, the dilatometry measurement will show $\Delta L_{(2\text{cm})}/L_{o(2\text{cm})} = \Delta L_{(3\text{cm})}/L_{o(3\text{cm})}$ (or $\Delta V_{(2\text{cm})}/V_{o(2\text{cm})} = \Delta V_{(3\text{cm})}/V_{o(3\text{cm})}$). However, if the surface effects such as oxidation and interface reaction form significant surface layer, L_s , in addition to the bulk swelling, the fractional length change in the shorter specimen will be larger than the longer specimen, $(\Delta L_{(2\text{cm})} + L_s)/L_{o(2\text{cm})} > (\Delta L_{(3\text{cm})} + L_s)/L_{o(3\text{cm})}$. The surface layer, L_s , contribute equally to both specimens since they are made from the same batch of material and tested in the same environment. Thus, the basis for using two different lengths (2 and 3 cm) specimens for the dilatometry is to differentiate between surface effects and volumetric swelling in the specimen.

In a separate experiment, the surface effect on the volume normalization was tested with a surrogate cerium metal (Alfa Aesar, stock no. 10139). Cerium metals are machined to the same specification as the plutonium specimens and tested in the dilatometry under the same conditions. Cerium metal is non-radioactive, and oxidizes rather easily after its surface is polished; it is an ideal candidate for validating surface

oxidation effects to dimensional normalization. Figure 2 shows the 2 and 3 cm length cerium metals held at 50°C under the helium atmosphere in the dilatometer. The shorter specimen shows larger $\Delta V/V_0$ change compared to the longer sample. This indicates that surface effects such as oxidation can cause the shorter sample to show larger volume changes when normalized to the initial volume as discussed above. For the enriched plutonium alloys in Figure 1, however, both 2 and 3 cm length specimens at 35°C show similar $\Delta V/V_0$ values indicating minimal surface contribution, and the measured volume change is therefore representative of the bulk swelling from the self-irradiation damage.

3.3 Physical interpretation of volumetric swelling

During the initial stage of aging at 35°C in Figure 1, the volume change follows approximately the inverse exponential-type of expansion. This behavior has been observed as an effect of radiation damage in materials [13, 14]. The amount of swelling in this case is approximately related to the number of Frenkel pairs that survive the radiation damage and subsequent recombination processes. The progressive accumulation of survivor vacancies provides an increasing number of alternate sites for the capture of self-interstitial plutonium atoms. As the density of these alternate sites increases, the rate of swelling thereby reduced. As discussed above, Chebotarev and Utkina also observed this initial transient stage using X-ray diffraction technique on plutonium alloys stored for 2 to 3 years [3]. Based on their results, the initial volume increase is due primarily to increase in lattice parameters. This initial stage is observed in the current work up to about 2 equivalent years of aging for samples at 35°C where the volume expansion becomes reduced. After the initial transient stage, the dilatometry

work shows almost linear volume expansion primarily caused by the helium in-growth. The helium that accumulates in plutonium immediately finds an unfilled vacancy and forms bubbles inside the crystalline matrix and along grain boundaries [5-7]. The dilatometry shows volume expansion at a steady rate with time primarily induced by a constant helium in-growth rate of ~41.1 appm per year, whereas the data of Chebotarev and Utkina do not [4]. This disagreement is caused by the differences in the measurement techniques. The dilatometry technique measures the changes in the bulk of the material. Thus, it is sensitive to length (volume) changes caused by both the lattice damage and the helium accumulation. Although the X-ray diffraction technique can measure the lattice parameter changes caused by the lattice damage, it is insensitive to the bulk swelling caused by the helium accumulation.

3.4 Volumetric swelling analysis

The contributors to the inverse exponential-type and linear volume swelling are changes in lattice parameters and the build-up of helium, respectively. Thus, the curves at 35°C are quite accurately represented by the combination of exponential [13] and linear growth equations of the form

$$\Delta V/V_0 = A [1 - \exp(-Bt)] + Ct \quad (2)$$

where A is the swelling value at saturation, B is related to the damage volume per time, C is the linear swelling rate by helium accumulation, and t is the time in years. The constants (A , B , and C), as determined from the curve fit, are given in Table 3, along with the He-to-vacancy association ratios calculated using the slope (C). This ratio can be approximated with the relationship [1]

$$\Delta V/V_0 \cong [He]/(R) \quad (3)$$

where R represents the He-to-vacancy ratio and the radiogenic helium concentration $[He]$ is given in atomic parts per million. The helium production rate is 41.1 appm (atomic parts per million) per year. This model describes the linear volume expansion induced by the formation of helium bubbles in the Pu alloy and the average number of helium atoms taking up the volume of a vacancy. The extracted He/vacancy ratios from the equation are 2.66 and 1.83 at 35°C indicating more than one helium atom per vacant lattice site, and the constant C represents the corresponding rate of volume swelling. This ratio should be one if each helium atom captured by bubbles brings with it a vacancy. Thus, helium atoms can stabilize vacancies and can have an important influence on the subsequent changes in plutonium properties.

Because Equation 2 represents the model of volumetric swelling, it does not describe the volume change with potential contributions from surface effects for samples at 50 and 65°C. Table 3 includes the constants from the curve fit to 50°C and 65°C for comparison to 35°C. With the exception of the specimen at 50°C (3 cm length), the constant A is larger and B is smaller than at 35°C, potentially caused by surface effects as discussed in Section 3.2. However, once the surface passivity is reached, the contribution from the surface oxidation or reaction layer to the volume change will be minimal and the linear expansion portion of the volume change curve can be analyzed with the linear growth portion in Equation 2. The passivated surface layer, estimated to be a few microns thick and contributed to the initial volume increase, will have minimal contribution to the volume increase by the helium in-growth. By this approach, the He-to-vacancy ratio can be extracted with the error primarily originating from the systematic ($\pm 0.10 \mu\text{m}$) and

curve fit uncertainty. The average ratio from all specimens is about 2.3. This He-to-vacancy ratio represents the helium density within the bubble. At this ratio, it is estimated that the pressure inside the helium bubble is almost off-set by the surface tension of the bubble formed in plutonium [1, 15]. If the ratio was larger, between 3 and 4, the significantly greater helium bubble pressure would cause the bubble to grow by dislocation loop punching [15]. At the current ratio, however, the swelling is estimated to be substantially less than 0.4 percent even after 100 years of aging. It is therefore important to examine void swelling mechanisms for any significant future density changes. While this ratio is corroborated by the observation from TEM and positron annihilation measurements [5-7], a significant variation exists between specimens, see Table 3. Even 2 and 3 cm length specimens at 35°C, which represent bulk changes, show different values of 2.66 and 1.83. Whether this large variation is due to a deficiency in the theoretical models (Eqs. 2 and 3) for analyzing data and computing He-to-vacancy ratio or due to the uncertainty of measurements is unclear at this time. More measurements and more detailed analysis are under way to better quantify the dimensional changes of plutonium alloys by aging.

3.5 Immersion density

In order to compare and validate the accelerated aging method, the density change in enriched alloys are measured and compared to naturally aged reference plutonium alloys. Comparison of density change observed in aged reference alloys and ^{238}Pu -enriched alloys to the 35°C dilatometry data is shown in Figure 3. The immersion density measurement on the reference and ^{238}Pu -enriched alloys showed initial densities of

15.795 and 15.78 g/cm³, respectively. The ages of reference samples range from 0.2 to 21 years. Both reference and enriched alloys show reasonable agreement in density changes over time. The density converted from the dilatometry at time = 0 is set to 15.795 g/cm³ to compare to density changes of the reference alloys. The trend in the density change converted from the dilatometry corresponds well to the immersion density during the initial (exponential) transient stage predominately caused by the lattice damage from the radioactive decay. Following the initial stage, the rate of density reduction becomes reduced as observed from the dilatometry. Both dilatometry and immersion density measurements show dimensional and density changes in enriched Pu alloys induced by the self-irradiation damage. However, no evidence of void swelling is yet observed.

4. Summary and Conclusion

The self induced damage of alpha decay process in plutonium is known to damage its crystal lattice and generate uranium and helium atoms. While the majority of displaced atoms quickly return to lattice sites, the remaining Frenkel-type defects and accumulated helium atoms drive microstructural evolution. Potential consequence of self-irradiation damage effects in plutonium, after a quiescent period, is a void swelling. We report initial dimensional changes observed from the in-situ dilatometry on ²³⁸Pu-enriched alloys supplemented by immersion density measurements on both enriched and reference alloys. Dilatometry shows the dimensional expansion from the accumulation of residual lattice damage and helium in-growth. We found reasonable agreement in the density change behavior between enriched and reference alloys from combined dilatometry and

immersion density measurements. At twenty-five equivalent years of aging, there is no evidence of void swelling in any of the materials. Dilatometry measurements show volumetric changes of 0.11 % and 0.12 % for 2 and 3 cm length samples at 35°C while showing very slight differences for specimens at higher temperature. Higher temperature measurements are potentially affected by surface effects such as oxidation and/or interfacial reaction and require further investigation. By modeling the volume swelling measured by the in-situ dilatometry, the average He-to-vacancy ratio from tested specimens is estimated to be around 2.3 indicating that the helium bubble pressure is approximately in equilibrium with the surface tension of the bubble formed in plutonium alloy.

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References

1. W.G. Wolfer, Los Alamos Sci., 26 (2000) 274.
2. S.S. Hecker and J.C. Martz, Los Alamos Sci., 26 (2000) 238.
3. H. Ullmaier, Mater. Res. Soc. Bull., 22 (4) (1997) 14.
4. N.T. Chebotarev and O.N. Utkina, Plutonium and Other Actinides, 1975, H. Blank and R. Lindner, (Eds.), Amsterdam, (1976) 559.
5. P.A. Sterne, J. van Ek, R.H. Howell, Comput. Mater Sci., 10 (1998) 306.
6. P. Asoka-Kumar, S. Glade, P.A. Sterne, and R.H. Howell, Plutonium Futures – The Science Conference, July 6-10, 2003, Albuquerque, NM.
7. A.J. Schwartz, M.A. Wall, T.G. Zocco, and W.G. Wolfer, Phil. Mag., 85, (2005) 479.
8. M.J. Fluss, B.D. Wirth, M. Wall, T.E. Felter, M.J. Caturla, A. Kubota, and T. Diaz de la Rubia, J. Alloys and Comp., 368 (2004) 62.
9. J.A. Lee, K. Mendelssohn, and D.A. Wigley, Physics Letters, 1 (1962) 325.
10. K. E. Dodson, O. H. Krikorian, M. S. Blau, W. L. Williams, J. A. Schmitz, D. A. Mew, P. J. Benevento, and B. B. Ebbinghaus, Lawrence Livermore National Laboratory Report, UCRL-ID-148262, May 6, 2002.
11. B.W. Chung, S.R. Thompson, C.H. Conrad, D.J. Hopkins, W.H. Gourdin, B.B. Ebbinghaus, Mater. Res. Soc. Proc., Symp. DD, Boston, MA, 2003.
12. H. A. Bowman, R.M. Schoonover, and M.W. Jones, J. of Res. Nat. Bur. Stand., 71C (1967) 179.
13. W. J. Weber, J. W. Wald, and H. Matzke, J. Nucl. Mater., 138 (1986) 196.

14. W.J. Weber, and F. P. Roberts, Nucl. Tech., 60 (1983) 178.
15. A. Arsenlis, W.G. Wolfer, and A.J. Schwartz, J. Nucl. Mater., 336 (2005) 31.

Table Caption

Table 1. Induction coupled mass spectrometric analysis is given for reference and enriched alloy. Standard deviations are given in parentheses. Elements below the minimum detection limit and undetectable from the analysis are denoted as “D” and “ND” in the table, respectively.

Table 2. Summary of length change and volume expansion of ^{238}Pu -enriched alloys after 25 equivalent years of aging.

Table 3. Values of constants from the curve fit (Eq. 2) to Figure 1. Equation 2, however, does not accurately represent dimensional changes at 50°C and 65°C due to potential contribution from surface effects (see text). He-to-vacancy ratios are calculated from the parameter C and Eq. 3 with the helium production rate of 41.1 appm per year. The major contributions to each ratio’s error are from the systematic ($\pm 0.10\ \mu\text{m}$) and curve fit uncertainty.

Figure Caption

Figure 1. The normalized volume changes for ^{238}Pu -enriched alloys tested under helium atmosphere at 35, 50, and 65°C. A pair of 2 and 3 cm length specimens was tested at each temperature. The time is represented as an equivalent time calculated by taking the measurement time and multiplying by the accelerating factor.

Figure 2. The normalized volume changes for cerium metals at 50°C under helium atmosphere. The comparison shows that surface effects such as oxidation cause the shorter sample to show a larger volume change when normalized to the initial volume.

Figure 3. Comparison of density changes between the enriched alloys from the dilatometry (35°C) to reference and enriched alloys from the immersion density.

Table 1.

Element	Minimum Detection Limit	Reference Alloy	Enriched Alloy	Units
V	9.2	43.9(23)	27.7(5)	appm
Cr	4.5	D	19.9(22)	appm
Fe	42	1729(126)	1702(139)	appm
Ni	4	365(8)	20(8)	appm
Ga	16	18984(67)	20302(473)	appm
Y	0.5	ND	87.3(64)	appm
Ta	0.3	ND	67.6(93)	appm
W	0.3	ND	1.7(0.1)	appm
²³⁴ U	0.2	3(1)	181(44)	appm
²³⁵ U	0.2	80(2)	9(2)	appm
²³⁷ Np	0.2	11(1)	66(1)	appm
²³⁸ UPu	0.001	0.013(5)	7.29(6)	at. %
²³⁹ Pu	0.05	91.80(10)	84.84(1)	at. %
²⁴⁰ Pu	0.001	5.85(5)	5.41(6)	at. %
²⁴¹ PuAm	1	0.169(5)	0.146(6)	at. %
²⁴² Pu	0.5	0.046(5)	0.055(4)	at. %

Table 2.

Sample Temp	Sample Length (nominal)	ΔL (μm, ± 0.10)	$\Delta V/V_0$ (%)
35°C	2 cm	7.42	0.11
35°C	3 cm	12.03	0.12
50°C	2 cm	13.40	0.20
50°C	3 cm	13.75	0.14
65°C	2 cm	14.91	0.22
65°C	3 cm	14.89	0.15

Table 3.

Sample Temp.	Sample Length (mm, ± 0.012)	A (10^{-4})	B	C (10^{-5})	He/vacancy ratio
35°C	20.009	7.45 ± 0.12	0.88 ± 0.04	1.55 ± 0.07	2.66 ± 0.28
35°C	30.009	6.71 ± 0.15	1.17 ± 0.09	2.25 ± 0.10	1.83 ± 0.13
50°C	20.026	14.92 ± 0.46	0.31 ± 0.01	2.12 ± 0.24	1.94 ± 0.31
50°C	30.015	7.80 ± 0.25	0.79 ± 0.07	2.51 ± 0.15	1.64 ± 0.14
65°C	20.023	18.59 ± 2.64	0.52 ± 0.20	1.35 ± 0.15	3.05 ± 0.56
65°C	30.025	10.89 ± 1.07	0.59 ± 0.01	1.49 ± 0.06	2.77 ± 0.30

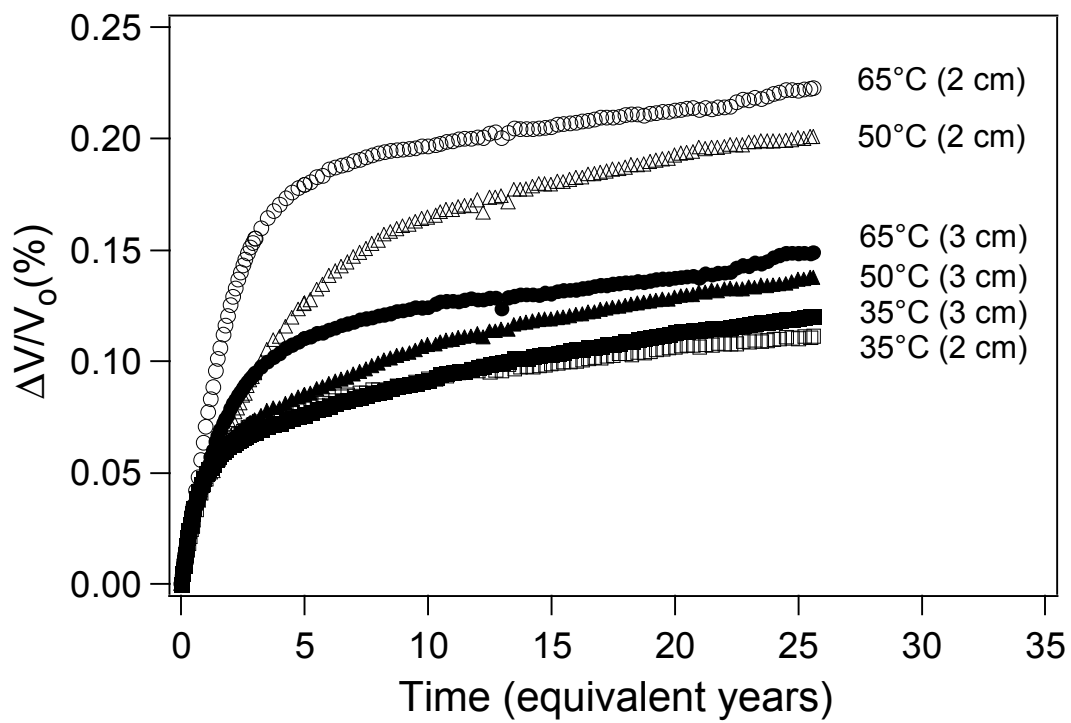


Figure 1

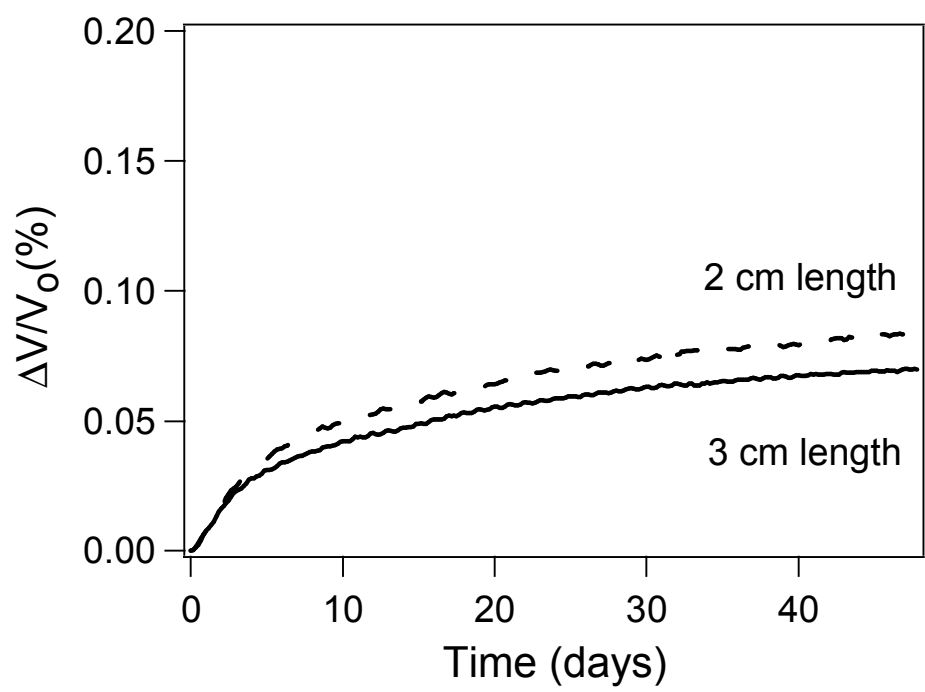


Figure 2

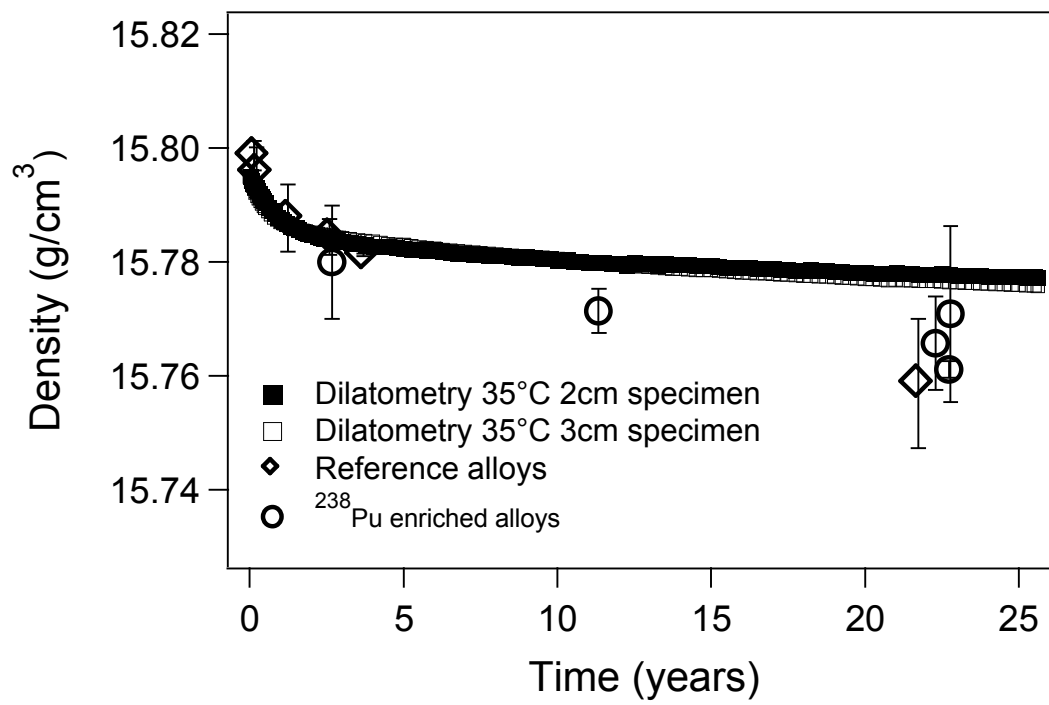


Figure 3